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Advanced characterization: aberration-corrected scanning transmission electron microscope (AC-STEM) capability and technique development

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Abstract

This work has started the process of extending nanometer-scale comprehensive microanalysis to the 3rd dimension by combining full x-ray spectral imaging with previously developed computed tomography techniques whereby we acquire a series of spectral images for a large number of projections of the same specimen in the transmission electron microscope and then analyze the composite computed tomographic spectral image data prior to application of existing tomographic reconstruction software. We have demonstrated a needle-shaped specimen geometry (shape/size and preparation method) by focused ion beam preparation and acquisition and analysis of a complete tomographic spectral image on a test material consisting of fine-grained Ni with sub-10 nm alumina particles.

Introduction

Microanalysis at nanometer to sub-nanometer length scales is critical to understanding the earliest aging-related changes in materials. In addition for many microstructures, a single 2D projection may be insufficient for a complete understanding of chemical changes due to aging and thus high-resolution 3D microanalytical techniques are needed. The goal is to develop operational competence and pushing to the practical resolution and sensitivity limits of the new instrumentation, in 3D, ultimately to NW-relevant materials systems. In order to develop this approach, however, we started with a test material consisting of fine-grained Ni with small alumina particles throughout the microstructure.

Approach

Computed tomography has become more or less routine for transmission electron microscopy but adding chemical signals to tomography experiments has not been extensively pursued due to poor x-ray collection efficiency--a problem solved with the new aberration-corrected scanning transmission electron microscope (AC-STEM) and its 0.7 sr x-ray detector array. Figure 1 shows a schematic of the AC-STEM and the main features of this instrument which make it suitable for this work: a high-brightness field-emission electron source (X-FEG); spherical aberration corrector on the probe forming optics (DCOR) and large solid-angle x-ray detector array (SuperX). This analytical electron microscope is 100 times better in terms of spatial resolution, sensitivity and signal acquisition time versus our next best system at Sandia purchased in 2000. We now have specialized specimen holders making tomography possible but the techniques for microanalytical tomography, by for example x-ray microanalysis, must be developed and tested. This includes the choice of relevant materials systems, optimization of specimen preparation, data acquisition and analysis. Sandia possesses the software for constructing the 3D model of a sample from a series of projection-images (computed tomography software) but we require development of techniques for routinely collecting chemical tomograms and reducing them to 3D models. In this report we will describe the foundational work needed to routinely perform 3D microanalysis on thin samples, including specimen preparation and data acquisition and analysis.

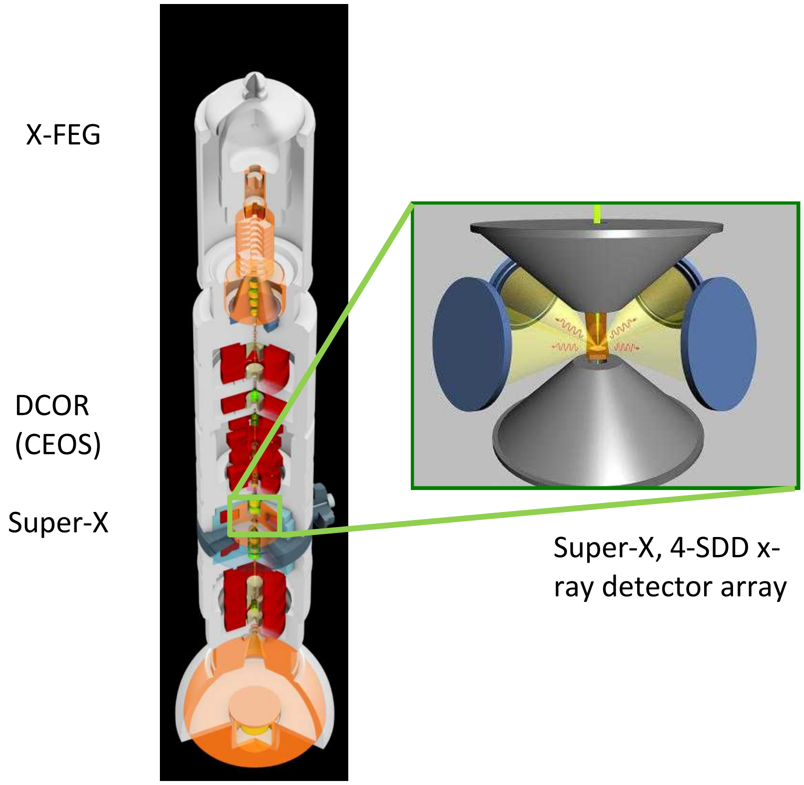

Figure 1. Schematic of the FEI Titan 80-200 with ChemiSTEM technology.

Results and Impacts

Foremost to this work is the need to develop a specimen geometry amenable to 3D microanalysis. Given that our materials aging problems often involve site-specific defects, we used the focused ion-beam tool to prepare a thin (200nm thick or less in projection) sample. Furthermore since the goat is to collect x-ray date for each of a number of different projections, a needle sample geometry was chosen, whereby a thin needle-shape sample is milled from a bulk material on top of a sub-100µm diameter post which fits in a special tomography holder into the TEM. This tomography holder can be rotated in one axis through 360° without shadowing any of the x-ray detectors thus maximizing our analytical sensitivity. Figure 2 shows the analytical geometry developed for this work. In this a brass pin has had its end milled in the FIB to be sharper and less likely to shadow the x-ray detectors in the ASC-STEM. Then a small piece of the material of interest (Ni-alumina) has been milled from the bulk, placed upon the tip and then thinned to a needle shape which is less than 50 nm in diameter at the tip. The pin is then placed in the specimen holder as shown in Fig. 2E which can then be inserted into the AC-STEM. Once in the AC-STEM, x-ray spectral images are acquired from a series of 29 projections from -90° to +90°. In practice we recorded a spectral image with 250 by 1000 pixels each with 2048 spectral channels at 1nm/pixel spatial resolution at a projection then rotated by 5° and then collected another spectral image until we had complete tomographic spectral image.

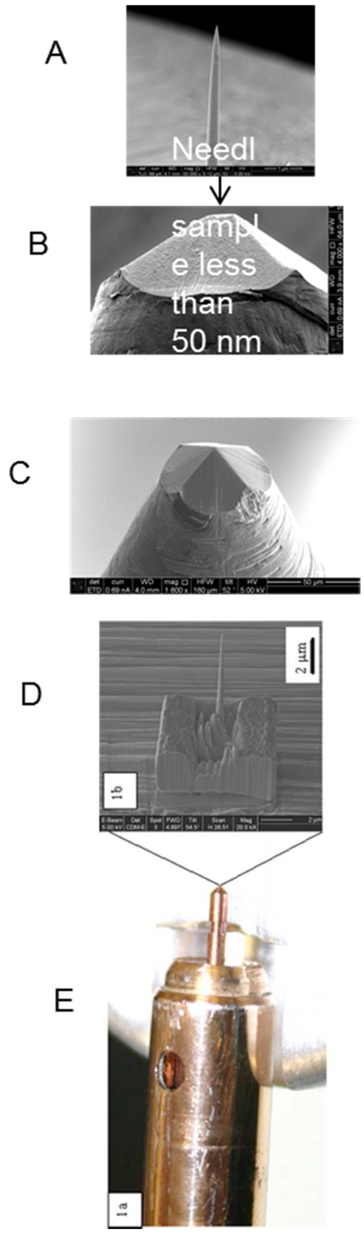

Figure 2. A. Final needle specimen with a tip less than 50 nm in diameter, supported by a brass pillar (B&C, different views) which has had its end milled in the FIB to be sharper, and that fits in the sample holder as shown in E.

The entire tomographic spectral image was then analyzed with Sandia's Automated eXpert Spectral Image Analysis (AXSIA) software [1,2] which results in a component spectrum and corresponding projection-series images. The projection images for each component then serve as input for the commercially available tomographic reconstruction package, Inspect3D from FEI Company. Figure 3 shows the work-flow for the data acquisition and analysis.

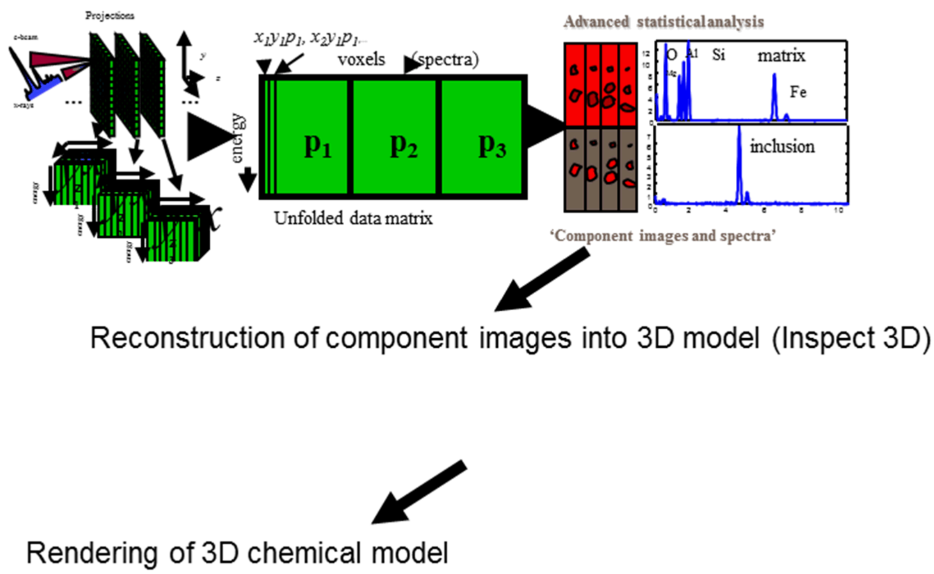

Figure 3. Workflow for tomographic spectral image acquisition and analysis.

The individual spectral images from each projection were combined into a single binary file for analysis by AXSIA which for this 14.8 billion data element file took 166 seconds. Figure 4 shows a subset of the results including the spectral shapes for Ni and Al-O as well projection image color overlays for three of the 29 projections.

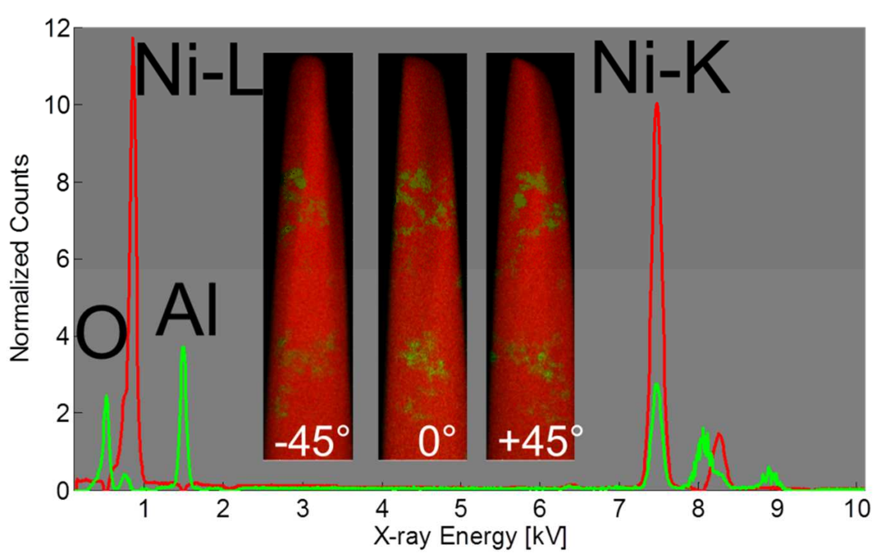

Figure 4. AXSIA results showing three projections (out of 29). Ni is red and Al-O is green. The small green regions in the images correspond to alumina particles. The underlying component images in the overlay form the input to the reconstruction software.

The component images (with corresponding chemistry, e.g., Al-O) have then been used as input into commercially available tomographic reconstruction software, Inspect3D from FEI Company. This software is typically used to perform 3D reconstructions from series of images such as bright-field TEM images. Such images are often contaminated with diffraction information from the crystalline materials in the sample. Such diffraction contrast can interfere with the reconstruction algorithms as they are expecting simple mass-thickness contrast. In contrast, the reduced full spectral information has very little diffraction contamination and thus conforms better to the reconstruction algorithm's assumptions. A preliminary 3D reconstruction from the tomographic spectral image is shown in Figure 5. Here we show only the alumina particles in the Ni needle.

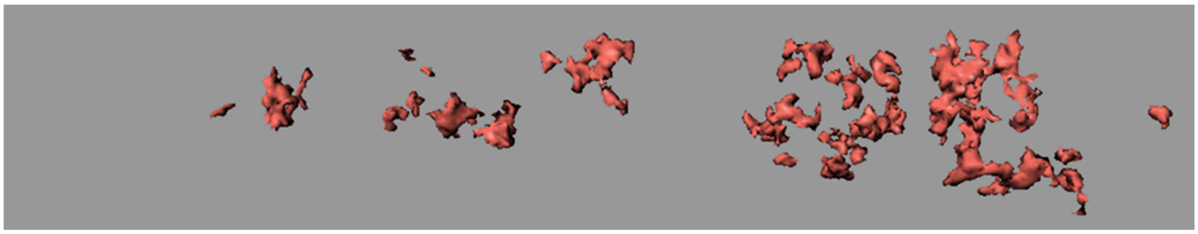

Figure 5. Reconstruction of the Ni/Alumina needle from the tomographic spectral image data. Individual particles are approximately 10nm in diameter.

Conclusions and Future Work

We have demonstrates a method of comprehensive 3D microanalysis on a Ni/Alumina test specimen at a resolution approaching several nanometers. Using computed tomography and full x-ray spectral images at each projection we were able to reconstruct the 3D elemental distribution for the sample. Future work will further develop these methods by direct application to materials of relevance to nuclear weapons and also by testing sensitivity to low levels of, for example, interfacial segregation.In order to test detection limits we will apply these methods to a specimen of known interfacial segregation.

Summary of Findings and Capabilities Related to Aging

A novel method of 3D microanalysis has been demonstrated which could be of relevance to studies of materials aging.

References

1. P.G. Kotula, M.R. Keenan, and J.R. Michael, "Automated Analysis of SEM X-Ray Spectrum Images: A Powerful New Microanalysis Tool," *Microscopy and Microanalysis* **9** [1] (2003) 1-17

2. P.G. Kotula, M.R. Keenan, and J. R. Michael, "Tomographic Spectral Imaging with Multivariate Statistical Analysis: Comprehensive 3D Microanalysis," *Microscopy and Microanalysis* **12**, 36-48 (2006)

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Administrative Addendum

Description	Quarter (1, 2, 3, 4)	Target Date (MM/DD/YY)	Complete?
Collect a chemical tomogram via x-rays on a test specimen	Q2-3	6/1/13	Yes
Develop 3D model and explore prospects for full 3D quantification and 3D detection limits	Q4	9/15/13	Yes, except detection limit issue

Milestones and completion status.

- **Milestone Status:**

In the Table above

- **Financial Leveraging:**

The work represented was paid for by:

Source	Dollar Amount ($k)
Enhanced Surveillance	50
Surveillance	[XX]
Systems	[XX]